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Report:

The discovery of the Invar effect [1], that is the anomalously low thermal expansion of $Fe_{64}Ni_{36}$ and $Fe_{72}Pt_{28}$ alloys over a wide range of temperature has stimulated many experimental and theoretical studies although no full understanding of the effect has been achieved so far. One of the most commonly accepted models is the so-called "2-γ"-state model first proposed by Weiss [2]. According to this model, iron can occupy two different states: A high-volume state and a low-volume state located at slightly higher energy. The low volume state can be thermally populated thus compensating for the thermal expansion. The Weiss model has been supported by first-principle calculations showing the existence of a high-spin (high-volume) to low-spin (low-volume) transition in Invar alloys as function of temperature [3]. However, no direct experimental evidence confirming the 2-γ state has been seen so far [4].

A recent reexamination of the Fe-Ni alloys magnetic properties within an *ab-initio* model has given a new impulse to the understanding of Invar effect. The model proposed by Shilfgaarde *et al.* [5] explains the volume anomaly by the non-collinearity of the iron- (and to a smaller extend of the nickel-) magnetic moments, the variation of the magnetic moments amplitude (as in the 2-γ state model) being of minor importance .

We report here the investigation of iron magnetism in the Invar alloy of Fe-Ni as function of pressure by X-ray emission spectroscopy (XES). The sample was a $Fe_{64}Ni_{36}$ 10 micron-thick foil. The Kβ emission line (3p→1s transition) was energy-analyzed by a 1-m Rowland circle spectrometer. The analyzer was a Si(531) spherically bent single crystal operating around the Bragg angle of 73.12° corresponding to the Fe Kβ energy (7057 eV).

The Fe Kβ line in $Fe_{64}Ni_{36}$ was measured in the DAC at 300 K as a function of pressure in the 0-20 GPa range. The two extreme pressures XES spectra, measured at 0 and 20 GPa, are shown in the left panel Fig. 1. The spectra are composed of a main line centered at 7057 eV followed by a satellite structure on the low energy side. As emphasized in comparable experiments [6], the intensity of this satellite peak can be related directly to the presence, and eventually magnitude, of a magnetic moment in the $3d^n$ ground state configuration of the sample 3d valence orbitals.

The information on the magnetic moment of the Fe atom can be extracted from the Fe Kβ XES spectra taking the difference between the spectrum measured at pressure P and the lowest pressure point (P=0). This is done after normalization to one of the integral of all the spectra. The resulting difference spectra (not shown here) are mainly composed of two peaks of opposite sign . These two structures grow in magnitude with increasing pressure. A quantitative analysis to extract the relevant magnetic information is done by determining the integral of the absolute value of these difference spectra ΔI. The pressure dependence -ΔI(P) is shown in the right panel Fig. 1. In the low pressure region below 5 GPa, the curve presents a sharp decrease followed by a plateau in the intermediate pressure region which extends up to about 10 GPa. At higher pressures, the intensity abruptly goes to zero around 12 GPa and remains there up to the highest measured pressure point around 20 GPa. This shows that two magnetic transitions take place in the 2-5 GPa and in the 10-13 GPa ranges respectively. Further information on the magnetism in Invar can be obtained comparing the Invar XES spectra with those previously measured in pure iron under pressure. As shown in Fig. 1, the pure iron and the Invar spectra at 0 GPa, and those at high pressure are almost identical. This confirms that: *i)* the Fe atom in the Invar alloy is in a high-spin state at zero pressure as it is in α-iron, and, *ii)* at high pressure (20 GPa), the Fe atom in the Invar alloy is in a non-magnetic state as it is in ε-iron. From the equivalence between the low and high pressure spectra of Invar and pure iron it is possible to scale -ΔI(P) into the iron magnetic moment in absolute units $\mu_{Fe}(P)$, as shown on the right axis of Fig. 1

The evolution of the local magnetic moment with pressure differs in many way from the theoretical predictions in the non-collinear model (see Fig. 1 right panel). Although the present study should still be completed with XES measurements on a finer pressure grid around 10 GPa and a temperature dependence investigation, the results reported here, strongly supports the model that the major contribution to the Invar effect comes from a high-spin to low-spin transition in the iron atoms.

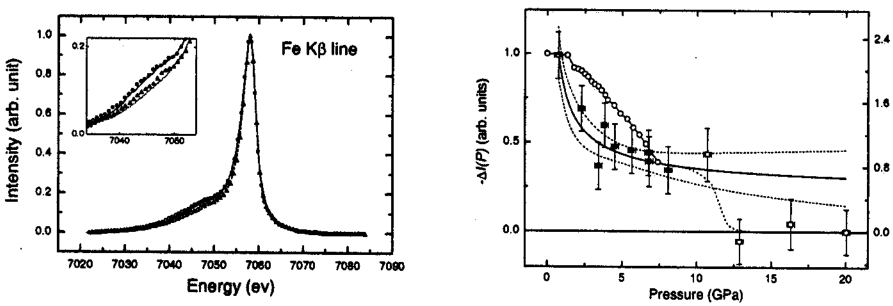

Figure 1 : Left panel : XES spectra measured at 0 (open circles) and 20 GPa (open triangles) in Fe-Ni Invar. As an element of comparison, XES spectra measured in pure Fe are also shown for both low-pressure α-Fe (solid line) and high-pressure ε-Fe (dash line) phases. A blow-up of the satellite region is represented in the inset. Right panel : Pressure dependence of the iron magnetic moment (solid squares) in the Fe-Ni Invar after analysis of the difference spectra. The right scale is deduced from the pure Fe XES data. The two-steps behavior strongly differs from the theoretical predictions (open circles)[5].

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